Slow and fast multi-photon ionization of clusters in strong XUV and X-ray pulses

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ABSTRACT

The photo-electron spectrum resulting from multi-photon absorption of an extended target, such as an atomic cluster or a large molecule, from an intense laser pulse with photon energies larger than the ionization potential of the atomic constituents is discussed. We develop an approximate analytical framework and provide simple analytical expressions for the shape of the photo-electron spectrum in the limit of sequential and parallel ionization, realized by long and short pulses, respectively. The width of the spectrum provides valuable information about the absorbed photons of the target in relation to its extension.

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1. Introduction

With the impressive development of attosecond laser pulse technology increasing attention has been paid to identify ultrafast processes on the attosecond scale which are promising and worthwhile to detect experimentally. Using attosecond pulses for making microscopic time measurements requires typically a pump–probe scheme to start and stop the clock. In attosecond streaking the system is pumped by an attosecond extreme-ultraviolet (XUV) pulse and probed by a few-cycle near-infrared (NIR) pulse. In early experiments [1,2] the final electron momenta were measured, in which the excitation time is encoded by the instantaneous vector potential of the NIR pulse. In another setup [3], the ion yield was recorded, whereby the *nonlinear* dependence of the tunnel ionization on the instantaneous electric field of the NIR pulse allows one to determine the initial time with subfemtosecond resolution. Using elliptical polarization attosecond time intervals for ionized electrons could be determined [4] provoking, as often, questions about the tunneling process itself. One of the most puzzling experimental finding using the XUV pump NIR streaking technique was the difference in ionization times for electrons released from different orbitals from a surface atom as well as an isolated gas-phase atom [5,6]. It could be shown that these attosecond time difference can be traced back to energy-dependent complex scattering phases in a time-independent picture [7] and that in general, the asymptotic phases are quite delicate to deal with [8]. In a broader context, it can be argued that for all processes which remain coherent until detected (i.e., no information is lost) time-

domain and energy-domain measurements are equivalent and related to each other by Fourier transform. In reality, this Fourier transform needs to take into account physical boundary conditions such as causality. Yet, it has been demonstrated that starting from the energy domain, i.e., by taking a series of frequency-resolved absorption spectra, one can indeed synthesize by an appropriate Fourier transform the time evolution of an electron density on an attosecond time scale [9].

Hence the question arises, for which kind of processes attosecond laser pulses allow to retrieve new and otherwise not obtainable experimental information. From the deliberations above it follows that we have to look for situations where information is lost in the energy domain on the way to the detector. This is clearly the case for all kinds of transient processes which are over once measured very much later at the detector. One example is the charging of a cluster or large molecule during illumination of a short intense laser pulse which remains hidden in conventional detection of charged (and often partially recombined) fragments at the detector. In this situation, an XUV attosecond pulse with well-defined time delay with respect to the strong laser pulse, could probe the transient ionic states *during* charging [10].

However, this is only one interesting non-mainstream application of attosecond pulses in extended systems. Here, we would like to draw attention to another phenomenon which is generic for intense XUV or X-ray pulses. For photon energies larger than the ionization potential multi-photon ionization in an extended system containing many atoms proceeds by almost simultaneous single-photon ionization of many atoms. This can lead to a transient electron cloud in the continuum of condensed-phase density which we will discuss in the following and contrast this ultrafast, parallel ionization in extended systems with its opposite, namely ultra-slow, sequential ionization. The latter was observed first in clusters

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and found to be well describable by Monte Carlo simulations [11]. We will discuss sequential and parallel ionization in the context of the so called Coulomb complexes (CC) [12]. They allow for an approximate description which leads to simple analytical expressions for the photo-electron spectra, despite the fact that we deal with many-particle systems.

2. General considerations

2.1. Photo-activation

If an extended target is illuminated by a laser pulse, typically the electrons bound to individual atoms or small molecules within the large target absorb a photon and become thereby photo-activated. The process of photo-activation can be described in a simple way by taking the appropriate photo-ionization cross sections σ_ω for the atom species contained in the cluster since multiple absorption of high-frequency photons by a single electron is very unlikely if the first photon absorbed puts the electron already into the continuum with respect to its mother ion. The single-photon absorption rate is proportional to the intensity $I(t)$ of the pulse,

$$\frac{dn(t)}{dt} = N_{at}\sigma_\omega \frac{I(t)}{\omega} \tag{1}$$

with $n(t)$ the number of electrons that have absorbed a photon up to time t, while N_{at} is the number of atoms. For a Gaussian pulse with duration T we can integrate Eq. (1) analytically to get

$$n(t) = \frac{N_{at}\sigma_\omega I_0}{\omega}\int_{-\infty}^{t} dt' \exp(-(t'/T)^2) \tag{2a}$$

$$= \frac{Q}{2}[1 + \mathrm{erf}(t/T)]; \tag{2b}$$

$$Q = N_{at}\sigma_\omega I_0 T\sqrt{\pi}/\omega, \tag{2c}$$

where Q is the total number of photons absorbed from the pulse. We use the variable Q to indicate that with Q photons absorbed the Coulomb complex becomes also Q-fold charged which we will need in the following. Clearly, taking a single-photon cross section is an idealization for several reasons. Yet, our main goal here is to achieve basic insight and keep the description as far as possible transparent and simple. Three snapshots before and during photo-activation are sketched in Fig. 1. Initially, the electrons (red) are bound by the ions (blue) which we will describe as a homogenuous positive background charge, see Section 2.2. An electron i, bound by an atom with energy E_b, which has absorbed a photon (green) of energy ω would be measured at the detector with the excess energy (atomic units are used unless stated otherwise)

$$E^* = \omega - E_b. \tag{3}$$

If the electron is released from a cluster atom, this excess energy is modified by a potential

$$\widetilde{E}^* = \omega - E_b + \widetilde{V}(\mathbf{r}). \tag{4}$$

Hence, coming out of a cluster, the electron will only be detected if its absorbed photon energy is sufficient to overcome the atomic binding energy E_b *and* the additional potential $\widetilde{V}$ due to the nearby ions and electrons in the system. The cluster potential $\widetilde{V}(\mathbf{r})$ depends on the position $\mathbf{r}$ of the electron and, most importantly, on the other electrons and consequently, the history of photo-activations in the system. The latter are determined by the duration of the laser pulse as we will discuss in Section 3. Before doing so we develop the simpler part of this potential, which is given by the ionic background.

2.2. The ionic background potential

Since we do not require details about the ions and their positions we approximate them by a jellium potential of constant charge density such that a cluster of radius R carries the charge Q. The potential created by this charge follows from

$$V(r) = \frac{3Q}{R^3}\int_0^R dr' \frac{r'^2}{|\mathbf{r} - \mathbf{r'}|} \tag{5}$$

which can be solved analytically using the partial-wave decomposition [13] of $|\mathbf{r} - \mathbf{r'}|^{-1}$ to give

$$V(r) = \begin{cases} -\frac{3Q}{2R} + \frac{Q}{2R}\left[\frac{r}{R}\right]^2 & \text{for } r \leqslant R \quad \text{(a)} \\ -\frac{Q}{r} & \text{for } R \leqslant r. \quad \text{(b)} \end{cases} \tag{6}$$

The depth of the potential $V_0 \equiv \frac{3}{2}Q/R$, cf. Fig.2, defines the energy scale of the extended system (note that this is not an absolute definition but depends on the photo-activation which generates Q charges, or from another perspective, deposits Q photons). The propagation of the photo-activated electrons can be done classically due to the high energies involved which can be measured by the excess energy $E^* \sim V_0$ relative to the harmonic curvature of the potential $\Omega = [Q/R^3]^{1/2}$, where $E^*/\Omega \sim (QR)^{1/2}$ which is of the order of 10^2 for 1000 photons absorbed by an extended system of a radius 1 nm ($\approx 20\,a_0$).

Interestingly, for a large and reasonable parameter space, we do not need to describe the motion of the ions in a simple, yet realistic approach. This can be seen be comparing the time scales of the electronic and the ionic motion. A typical time of the electron motion (with the order of the kinetic energy given by V_0) is

$$\tau_{el} = \frac{R}{\sqrt{2V_0}} = \sqrt{\frac{R^3}{3Q}}, \tag{7}$$

which scales with the period of the harmonic potential (6a). The ions move due to Coulomb explosion whereby the surface ions would be the fastest. Estimating the time it would take to change the radius of the ionic background by δR one gets [14]
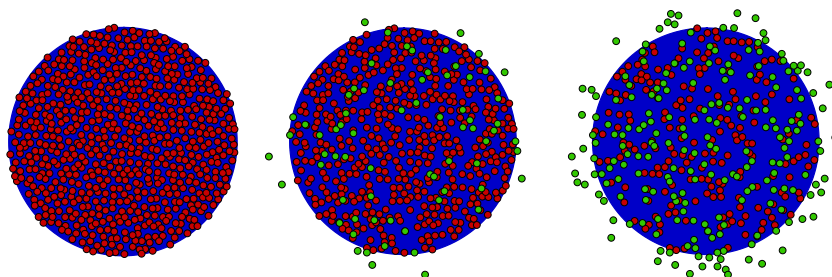
Fig. 1. Activation of photo-electrons in a Coulomb complex: bound electrons (red), activated electrons (green), ionic background potential (blue) for three different instances of time: $t \to -\infty$ (left), $t = -T/2$ (middle), and $t = +T/2$ (right). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)